Local Refractive Index Measurements at Low Temperatures using Photonic Crystal Cavities

Janik Wolters,[1,*] Niko Nikolay,[1] Max Schoengen,[2] Andreas W. Schell,[1] Jürgen Probst,[2] Bernd Löchel,[2] and Oliver Benson[1]

[1] *Nano-Optics, Institute of Physics, Humboldt-Universität zu Berlin, Newtonstr. 15, D-12489 Berlin, Germany*
[2] *Department for Micro- and Nanostructured Optical Systems, Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Albert-Einstein-Str. 15, D-12489 Berlin*

Photonic crystal cavities have a wide range of applications in physics today. Here we demonstrate a method to use the narrow resonances of photonic crystal cavities to measure the temperature dependence of the refractive index of gallium phosphide in a temperature range between 5 K and near room temperature at a wavelength of about 605 nm. On one hand, this is an essential step for the design of GaP photonic crystal structures for quantum technology applications. On the other hand, this demonstrates how photonic structures can be utilized to locally determine the optical properties of semiconductor materials in attoliter volumina.

Photonic crystal (PC) nano cavities have increasing importance for quantum optics, photonics and sensing applications[1]. The main reason for this is their ability to confine the electromagnetic field in volumina comparable to the cubic wavelength and thus to strongly enhance the light-matter interaction[2,3]. Using PC cavities not only the Purcell enhancement of single quantum emitters could be shown[4–6], but also the strong coupling regime of cavity quantum electrodynamics was reached with single quantum dots in PC cavities[7–9]. With such systems, lasing oscillations of a single quantum dot[10] and the ultrafast all-optical switching by single photons could be demonstrated recently[11]. For most of these experiments it is mandatory to cool the system to liquid helium temperatures, where the emission properties such as emission rate and coherence of most quantum emitters improve drastically[12]. However, at cryogenic temperatures also the optical properties of the photonic crystal slab material change dramatically, leading to a shift of the resonance wavelength and quality factor Q[13]. In order to fabricate complex integrated quantum circuits[3,7,14] the individual elements such as cavities have to be designed as precisely as possible. Therefore, design and material properties should be known a priori. One crucial aspect is the on-demand fabrication of a PC cavity with a specific Q and resonance at 5 K temperature. The resonance shift from room temperature to 5 K has two contributions: the change of the refractive index[15] and the change of the geometry due to thermal expansion[16].

In this letter we investigate GaP photonic crystal structures, operating in the visible[4,17]. In this material thermal expansion can be neglected and the only relevant process is the change of the refractive index n[15,16]. We quantify n precisely over a wide temperature range by measuring the resonance of photonic crystal cavities operating at a wavelength of about 605 nm in the temperature range between 5 K and near room temperature. Our method can be extended to any wavelength within the band gap of the material. We show that not only the resonance wavelength shifts by several nm, but also the Q-factor may increase by a factor of two if the PC is cooled to 5 K.

First, we designed 55 nm thick free-standing photonic crystal slabs (lattice constant 209 nm) with so-called L3 cavities[2] using FDTD simulations (*Lumerical*). These cavities are formed by three missing holes in a triagonal lattice. They support several spectrally narrow modes of high quality factors[18]. The electromagnetic field of the fundamental mode is concentrated mainly within GaP on about seven unitcells, corresponding to a material volume of 14 attoliter (see Fig. 1). The emission profile of this fundamental cavity mode is polarized perpendicular to the cavity axis[19], a feature that we will use later on to measure the cavity resonance. The spectral position and Q-factor of the mode depends not only on the geometry (lattice constant, hole radius, slab thickness), but as mentioned also strongly on the refractive index of the PC material.[13]

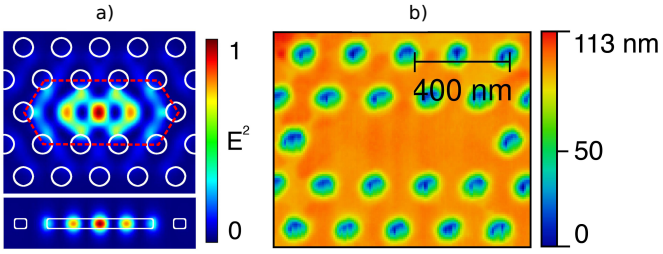

FIG. 1. (Color online) a) Simulated (FDTD) electric field profile in the slab center and cross-section of the fundamental mode of the cavity. The dashed red line indicates the seven unitcells in which the field is concentrated. b) Atomic force micrograph of the L3 cavity in GaP. The lattice constant is 209 nm.

For example, changing the refractive index of the PC, which has a volume of about 100 attoliter by about 0.01% corresponds to a wavelength shift by (54 ± 1) pm, which can be detected by a spectrograph and is relevant for experiments with high-Q cavities.[7] FDTD simulations unravel that in the studied regime the change of the resonance with the refractive index does not depend on the

thickness of the photonic crystal slab (Fig. 2). Thus, a change in the resonance wavelength is a very precise measure for changes in the refractive index, even if the exact geometry is not known.

We fabricated the designed structures by electron beam lithography from 59 nm thick heteroepitaxial gallium phosphide (GaP) layers deposited on Si(100) substrates.[20–22] The structures were transferred to the GaP layer by dry-etching with BCl_3 and subsequent removal of the underlying Si layer by isotropic dry-etching with SF_6. After processing, the quality and geometric precision of the structures were measured using scanning electron and atomic force microscopy (AFM). Fig 1b) shows an AFM image of the fabricated structure. To

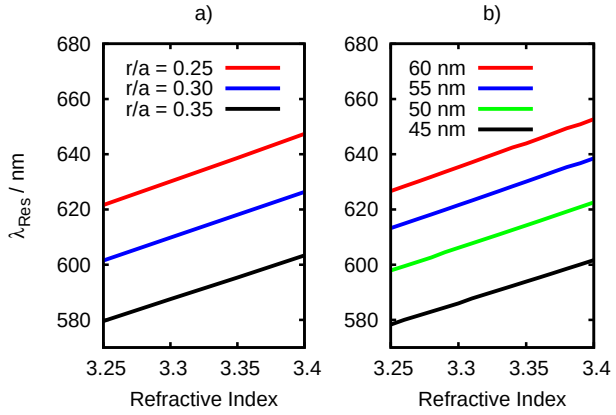

FIG. 2. (Color online) Simulated dependence of the first cavity resonance wavelength on the refractive index for various hole radii (a) and slab thicknesses (b). The behavior is nearly linear and the slope does not depend on the thickness of the slab or on the hole diameter in the studied regime.

control the cavity temperature within a wide range, the sample was mounted on the cold finger of a continuous flow He-cryostat. By changing the He-flow and additional heating the temperature could be precisely regulated in the range between 5 K and 300 K. In parallel, the spectral position of the cavity resonance was measured. Usually, the intrinsic material fluorescence under incoherent excitation is analyzed with a spectrograph for this purpose[23]. Here, this method could not be applied, as the excitation of fluorescence requires relative strong lasers (in the order of 10 μW), which would unavoidably heat the PC cavity. To avoid this heating, we analyzed the reflected light in the crossed polarization scheme[24]. In this scheme only very low illumination power is needed ($P < 100$ nW). By doubling the power and comparing the position of the cavity resonance we could guarantee that the radiation has negligibible influence on the cavity temperature ($<$ 1 K).

In detail, the cavity is illuminated with a vertical polarized collimated white light beam from a super continuum source (*NKT Photonics SuperK*) focused on the cavity by a microscope objective with a numerical aperture of 0.9 (*Mitutoyo*), which is placed inside the insulation vacuum of the cryostat. Importantly, the axis of the cavity is rotated by 45° with respect to the incident polarization. The reflected light is collected through the same objective lens, but only the horizontal polarized component of the reflected light is detected with a 500 mm spectrometer (*Acton SpectraPro 500i*) in a confocal configuration. A sketch of the setup is shown in Fig. 3. If

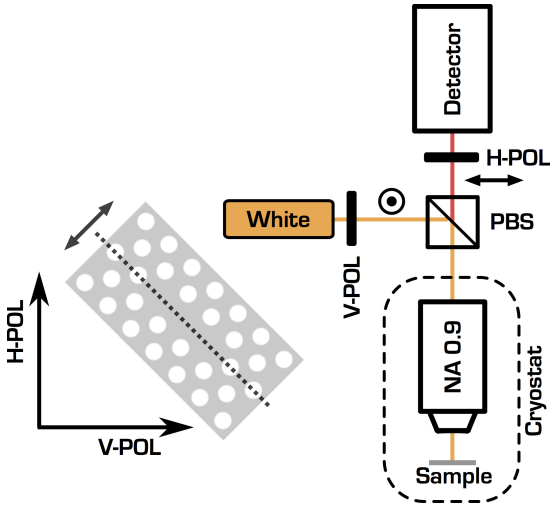

FIG. 3. (Color online) Sketch of the optical setup to measure the cavity resonance in the cross polarized detection scheme. The sample with the PC cavities is placed in the vacuum of a continuos flow He-cryostat to regulate the temperature. Through an objective lens with high numerical aperture the PC cavity is illuminated with vertical polarized white light (≈100 nW) from a supercontinuum source. The horizontal polarized component of the reflected light is detected in a confocal scheme by a spectrograph. The cavity axis is rotated by 45 degree with respect to the polarization basis. Thus, the polarization of the reflected light is slightly rotated at the cavity resonance. See text for details.

the incident light is not in resonance with the cavity, the beam is simply reflected and can not be detected by the spectrometer. If the light is in resonance with the cavity, there are two effects which slightly rotate the polarization and therefore can be detected on the spectrometer. On one hand, a small fraction of the resonant light will couple into the cavity and is not reflected. As this light is polarized perpendicular to the cavity axis, i.e. not parallel to the incident vertical polarization, the remaining part of the light which is reflected has a small horizontal polarized component. On the other hand, the light that couples into the cavity will be reemitted later. As the cavity mode is polarized along the preferential axis of the cavity the reemitted light reaches the spectrometer, too. This light is phase shifted with respect to the directly reflected one and interferes on the detector of the spectrometer with the latter. As a result of these

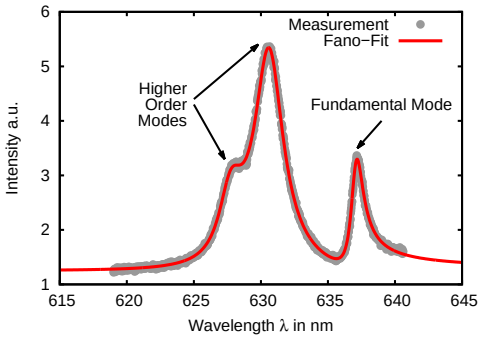

FIG. 4. (Color online) Example of the measured spectra and the corresponding fit with three Fano resonances at (637.03 ± 0.01) nm, (630.59 ± 0.01) nm and (627.68 ± 0.01) nm of an L3 cavity with hole radius $r = 50$ nm. The spectrum was taken at a temperature of 300K.

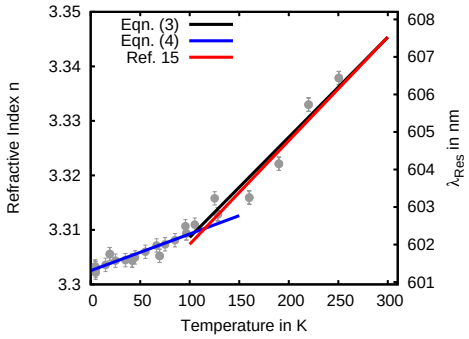

FIG. 5. (Color online) The measured cavity resonance wavelength λ_{Res} (right axis) and the calculated refractive index of GaP (left axis) between 5 K and 300 K. The data above 100 K are in good agreement with Ref.[15].

two interfering components, the cavity modes appear as Fano-resonances of the form:

$$F(\lambda) = A_0 + F_0 \frac{[q + 2(\lambda - \lambda_0)/\Gamma]^2}{1 + [2(\lambda - \lambda_0)/\Gamma]^2}, \qquad (1)$$

where A_0 is an offset, F_0 is the amplitude of the resonance, λ_0 is the resonance wavelength, $\Gamma = \lambda/Q$ the normalized resonance width and q the ratio of the amplitudes of the two paths[25]. Fig. 4 shows an example spectrum of a cavity with a hole radius of 50 nm and the fundamental resonance at about 637 nm, taken at room temperature. The spectrum can be perfectly fitted with three Fano-resonances. The fundamental mode supports the highest Q-factor of $Q = 580 \pm 50$. With the value from Ref.[26] for the refractive index at $T = 300$ K the slab thickness is estimated according to Fig. 2 to be about 55 nm.

In the following, the resonance wavelength of the fundamental mode of a cavity with hole radius $r = 63$ nm fabricated on the same sample was measured at various temperatures between 5 K and near room temperature. We calculated the refractive index n from the change of the resonance wavelength according to the relation

$$n(T) = n_{300K} + [\lambda(T) - \lambda_{300K}] \cdot a_{55}, \qquad (2)$$

with the refractive index at room temperature $n_{300K} = 3.34537$ from Ref.[26], the measured resonance wavelength at room temperature $\lambda_{300K} = (608.20 \pm 0.01)nm$ and the slope $a_{55} = 1/(163.36 \pm 0.01)$nm gained from the FDTD simulations for a slab thickness t of 55 nm (Fig. 2). The resulting data and fitted curves are shown in Fig. 5. Above 100 K the refractive index changes linearly according to

$$n(T) = 3.290 \pm 0.001 - (180 \pm 20) \cdot 10^{-6} \cdot T/\mathrm{K} \qquad (3)$$

This is in very good agreement with the already known values from Ref.[15]. Interestingly, the slope decreases at a temperature of $T \approx 100$ K and the refractive index follows

$$n(T) = 3.302 \pm 0.001 - (67 \pm 7) \cdot 10^{-6} \cdot T/\mathrm{K} \qquad (4)$$

We also tried to fit the entire dataset shown in Fig. 5 with an exponential function, but the two slopes model represented by Eqns. (3-4) is clearly more accurate. The change in the slope at about 100 K is qualitatively in agreement with older measurements of the low frequency dielectric constant from Ref.[27].

According to FDTD simulations the Q-factor is almost unaffected by a small change of the refractive index. Surprisingly, we find experimentally that the decrease of the refractive index is followed by an increasing Q factor (Fig. 6). Starting with a value of 560 ± 50 at room-temperature, the Q-factor of the fundamental mode increases linearly with decreasing temperature. The highest measured Q-factor of 1150 ± 50 is more than two times higher than the initial value at room-temperature.

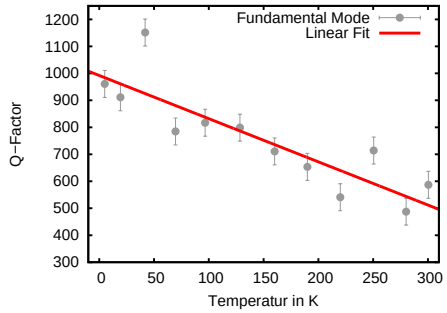

FIG. 6. (Color online) The measured Q-factor of the fundamental cavity resonance increases with decreasing temperatures. The highest measured value of $Q = 1150$ at $T = 42$ K is about two times bigger than the value at room temperature.

This behavior is probably attributed to reduced material absorption[28] at low temperatures and underlines once more how important it is to consider not only the desired operation wavelength, mode volume and Q-factor, but also the operation temperature of photonic crystal cavities.

In conclusion, we demonstrated how the narrow resonance of a GaP photonic crystal cavity shifts due to cooling. From this data we precisely calculated the refractive index of GaP in the ultrasmall cavity volume over the whole temperature range from near room temperature to $T = 5$ K. This knowledge is of great importance for the design and testing of PC structures and might also be used for novel PC applications. For example, the presented method may be used to determine the exact local cavity temperature. This is of great importance if quantum emitters are coupled to the cavity, as the emission properties of such emitters strongly change with the temperature. In most previous experiments the temperature of the cold finger of the cryostat was given as a temperature reference. However, the cavity structure may be weakly coupled thermally to the cold finger and excitation light may locally heat the cavity. This is particularly an issue when studying under-etched photonic crystal membrane cavities and diamond defect centers as emitters, which require a relatively large excitation power[4,29]. Finally, the index of refraction can be measured via our method for any material where fabrication of a resonant photonic structure is possible.

This work was supported by the DFG (BE2224/9). J. Wolters acknowledges funding by the state of Berlin (Elsa-Neumann). We thank H. Döscher and T. Hannappel for providing GaP on Si wafers.

* Electronic mail: janik.wolters@physik.hu-berlin.de

[1] K. J. Vahala, *Nature* **424**, 839-846 (2003).
[2] Y. Akahane, T. Asano, B. S. Song, S. Noda, *Nature* **425**, 4-7 (2003).
[3] J. L. O'Brien, A. Furusawa, J. Vuckovic, *Nature Photon.* **3**, 687-695 (2009).
[4] J.Wolters, A. W. Schell, G. Kewes, N. Nüsse, M. Schoengen, H. Döscher, T. Hannappel, B. Löchel, M. Barth, O. Benson, *Appl. Phys. Lett.* **97**, 141108 (2010).
[5] D. Englund, B. Shields, K. Rivoire, F. Hatami, J. Vuckovic, H. Park, M. D. Lukin, *Nano Lett.* **10**, 3922-6 (2010).
[6] D. Englund, A. Faraon, B. Zhang, Y. Yamamoto, J. Vuckovic, *Opt. Express* **15**, 5550-8 (2007).
[7] D. Englund, A. Faraon, I. Fushman, N. Stoltz, P. Petroff, J. Vuckovic, *Nature* **450**, 857-61 (2007).
[8] A. Faraon, A. Majumdar, D. Englund, E. Kim, M. Bajcsy, J. Vuckovic, *New J. Phys.* **13**, 055025 (2011).
[9] T. Yoshie, A. Scherer, J. Hendrickson, G. Khitrova, H. M. Gibbs, G. Rupper, C. Ell, O. B. Shchekin, D. G. Deppe, *Nature* **432**, 9-12 (2004).
[10] M. Nomura, N. Kumagai, S. Iwamoto, Y. Ota, Y. Arakawa, *Nature Phys.* **6**, 279-283 (2010).
[11] T. Volz, A. Reinhard, M. Winger, K. J. Hennessy, E. L. Hu, arXiv:1111.2915v1.
[12] F. Henneberger, O. Benson *Pan Stanford Publishing* (2009).
[13] J. D. Joannopoulos, S. G. Johnson, J N. Winn, R. D. Meade, *Princeton University Press* (2008).
[14] O. Benson, *Nature* **480**, 193-199 (2011).
[15] A. N. Pikhtin, V. T. Prokopenko, V. S. Rondarev, A. D. Yas'kov, *J. Appl. Spectroscopy* **27**, 1047-1052 (1977).
[16] G. A. Slack, S. F. Bartram, *J. Appl. Phys.* **46**, 89 (1975).
[17] K. Rivoire, A. Faraon, J. Vuckovic, *Appl. Phys. Lett.* **93**, 063103 (2008).
[18] R. Chalcraft, S. Lam, D. OBrien, T. F. Krauss, M. Sahin, D. Szymanski, D. Sanvitto, R. Oulton, M. S. Skolnick, M. Fox, D. M. Whittaker, H.-Y. Liu, M. Hopkinson, *Appl. Phys. Lett.* **90**, 241117 (2007).
[19] M. Barth, J. Stingl, O. Benson, *Appl. Phys. Lett.* **93**, 021112 (2008).
[20] H. Döscher, T. Hannappel, B. Kunert, A. Beyer, K. Volz, W. Stolz. *Appl. Phys. Lett.* **93**, 172110 (2008).
[21] H. Döscher, and T. Hannappel, *J. Appl. Phys.* **107**, 123523 (2010).
[22] H. Döscher, O. Supplie, S. Brückner, T. Hannappel, A. Beyer, J. Ohlmann, K. Volz, *J. Crystal Growth* **315**, 16-21 (2011).
[23] M. Barth, J. Kouba, J. Stingl, B. Löchel, O. Benson, *Opt. Express* **15**, 17231-40 (2007).
[24] M. Galli, S. L. Portalupi, M. Belotti, L. C. Andreani, L. O'Faolain, T. F. Krauss, *Appl. Phys. Lett.* **94**, 071101 (2009).
[25] U. Fano, *Phys. Rev.* **124**, 1866-1878 (1961).
[26] Landolt-Börnstein **41A1b**: Group IV Elements, IV-IV and III-V Compounds. Part b - Electronic, Transport, Optical and Other Properties, *Springer-Verlag* (2002).
[27] G. Samara, *Phys. Rev. B* **27**, 3494-3505 (1983).
[28] P. Dean, D. Thomas, *Phys. Rev.* **150**, 690-703 (1966).
[29] J. Wolters, G. Kewes, A. W. Schell, N. Nüsse, M. Schoengen, B. Löchel, T. Hanke, R. Bratschitsch, A. Leitenstorfer, T. Aichele, and O. Benson, *Phys. Stat. Sol. (b)* to be published DOI: 10.1002/pssb.2011001156 (2012).